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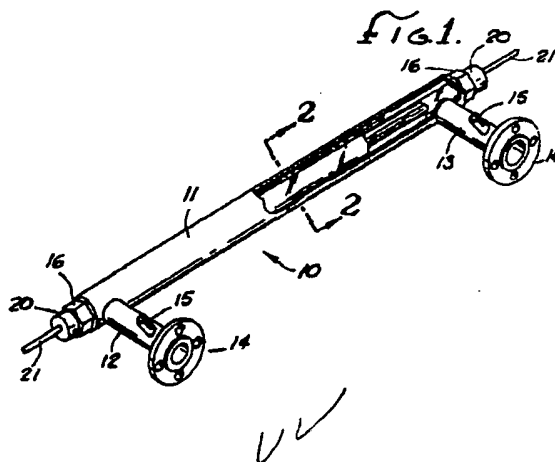
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(54) **Apparatus and method for conditioning water.**

(57) A method and apparatus for conditioning water by photochemically oxidizing organic materials in the water to reduce the Total Organic Carbon. The apparatus includes an elongated cylinder (11) with a water inlet (12) on one end and an outlet (13) on the other end and a quartz sleeve (17) concentrically positioned inside the cylinder (11) for forming an annular oxidizing chamber along the length of the cylinder (11) from the inlet (12) to the outlet (13). An ultraviolet light lamp (19) is positioned within the quartz sleeve (17) and produces 185nm ultraviolet light for oxidizing the organic material to carbon dioxide. A system is disclosed for reconditioning ion exchange resin columns (30, 31) by circulating water therethrough and through the TOC reduction unit to rinse organic materials from the resin columns (30,31) and convert the organic materials to carbon dioxide. Another system discloses the use of the TOC reduction unit and a deionization unit for maximizing the quality of water reclaimed in an industrial process.



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APPARATUS AND METHOD FOR CONDITIONING WATER

This invention relates to a method and apparatus for conditioning water and, in particular, but not limited to, for reducing the organics in an ion exchange resin column.

The water used in laboratory experiments and in many industrial processes, such as the manufacture of electronic components and computer chips, cooling for nuclear reactors, and the like, must be of as high a purity as possible or at least the water must be devoid of those impurities which adversely affect the laboratory experiments or industrial processes. Numerous types of filters of varying efficiencies have been developed for removing objectionable particulate matter. Ion exchangers of various types have been used for deionizing the water. Various additives and processes have been used for reducing or destroying the bacteria content of the water. However, the effective removal of organic impurities from the process water generally has been unsatisfactory. The organic impurities, usually referred to as Total Organic Carbon ("TOC") or Total Oxidizable Carbon, may result from any number of sources such as decaying vegetation, industrial solvents, oil, ion exchange systems, etc. While certain levels of TOC in water may be acceptable in many processes, there are processes in which it is detrimental or even unacceptable. Moreover, the presence of high levels of TOC may interfere with other water conditioning processes.

One useful function of the present invention, as will be described more fully below, is in conditioning or reconditioning ion exchange resins used in water purification systems. An ion exchanger is used for the deionization of water, namely, in removing the objectionable anions and cations from the water. There are various types of ion exchangers that use synthetic resins (polymers) of various types to produce the desired deionization. The resin forms a bed of "plastic sand" in a vessel through which the water is passed for treatment and, because of the shape, the ion exchanger is often referred to as a "resin column". After a period of use of a resin column, the resin must be regenerated or discarded if it cannot be regenerated effectively, and in any event after numerous regeneration cycles it must be replaced. For regeneration, the appropriate type chemicals are passed through the resin column to reverse the chemical processes which accomplished the deionization whereby the resin is returned to a composition which is again capable of accomplishing the desired deionization. During normal use, resin columns also accumulate organic impurities which tend to coat the resin and reduce the effectiveness

of the resin in deionizing the water. The chemicals used in regenerating the resin for returning its deionizing capability normally do not significantly reduce the organics accumulated on the resins. In some instances, relatively pure water is passed through resin columns in an attempt to rinse the organic impurities from the resin column but this consumes substantial quantities of water.

By the present invention there is provided an apparatus and method for substantially reducing or eliminating the Total Organic Carbon (TOC) in water through the intense exposure of the water to ultraviolet light of a specific wavelength for converting the organic impurities to carbon dioxide and other harmless oxidation products which then dissolve in the water. More specifically, an object of this invention is to provide a method and apparatus for exposing water to ultraviolet light having a wavelength of substantially 185nm (nanometers) for converting organic impurities in the water to primarily carbon dioxide.

Another object of this invention is to provide a method and apparatus for use in reconditioning ion exchange resin columns to remove the organic impurities therein by a closed-loop circulation of water through the resin column and an apparatus for intensely exposing the water to substantially 185nm ultraviolet light to convert the organic impurities to primarily carbon dioxide which in turn is removed by the resin, until substantially all of the organic impurities have been rinsed from the resin and reduced to carbon dioxide. A further object of this invention is to provide such a method and apparatus wherein subsequent to exposing the water to primarily 185nm ultraviolet light to reduce the organic impurities, the water is directly usable in a laboratory test or industrial process requiring minimal TOC levels in that water. A still further object of this invention is to provide a water treatment system wherein water that is reclaimed in an industrial process is recirculated through the apparatus of this invention for removing organics to allow reuse of the water.

Other and more detailed objects and advantages of this invention will be apparent to those skilled in the art from the following description and the accompanying drawings, wherein:

Fig. 1 is a perspective view of a preferred embodiment of the ultraviolet light apparatus of this invention;

Fig. 2 is an enlarged sectional elevation view of the apparatus of Fig. 1 taken on the line 2-2;

Fig. 3 is a schematic illustration of a system using the apparatus of Figs. 1 and 2 for TOC reduction of ion exchange resin columns; and

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Fig. 4 is a schematic illustration of a system using the apparatus of Figs. 1 and 2 for TOC reduction in water used in an industrial process for allowing reuse of the water.

Referring now to the preferred embodiment of the apparatus of this invention shown in Figs. 1 and 2, the TOC reduction unit, generally designated 10, includes an elongated cylinder 11 with a laterally extending pipe 12 mounted on the side at one end of the cylinder 11 and a second laterally extending pipe 13 mounted at the other end of the cylinder 11. The pipes 12 and 13 communicate with the interior of cylinder 11 to form an inlet and an outlet, respectively, to and from the cylinder. Each pipe 12 and 13 is provided with a means for connecting the pipe into a water circulating system and such means may comprise a flange 14 of a conventional type. Further, each pipe 12 and 13 includes a branch pipe 15 for the convenient connection of meters, sampling equipment, or the like for monitoring the properties of the water flowing into and out of the unit 10, or for the connection of fittings to allow the addition of oxygen or other oxidation enhancing additives to the water flowing into the cylinder 11.

Each end of the elongated cylinder 11 is provided with an axially extending threaded fitting on which a compression nut 16 may be threadably mounted. A quartz tube or sleeve 17 of a quality to transmit 185 nm light is positioned in the cylinder 11 and extends out each end for engagement by the compression nut 16 to connect and seal the quartz sleeve to the cylinder 11 in a conventional manner. By this conventional arrangement an annular shaped fluid chamber 18 is formed between the quartz sleeve 17 and the cylinder 11 for the passage of the water from inlet pipe 12 to outlet pipe 13 through the entire length of the cylinder 11. The mounting of the quartz sleeve 17 to the cylinder 11 by the compression nut 16 on each end results in a tubular passage through the length of the unit 10, which passage is opened at both ends. An ultraviolet light lamp 19 is positioned in the tubular passage formed by quartz sleeve 17 and electric lamp sockets 20 are adapted to be connected to each end of the lamp 19. There are a number of different types of conventional connections for the end of ultraviolet lamps in arrangements similar to this structure but it is preferred that the lamp socket 20 be of a size, shape and material to support the lamp 19 from each end and substantially center the lamp 19 within the quartz sleeve 17. By applying appropriate electrical power through wires 21 connected to each lamp socket 20 the ultraviolet light lamp 19 may be illuminated.

While the above described structure is similar to many conventional devices for the ultraviolet treatment of water and other fluids, there are some details of construction that are preferred for the TOC reduction unit 10 of this invention. Specifically, it has been found that for a presently conventional ultraviolet lamp of the low pressure type the fluid chamber 18 is preferably of a relatively narrow radial thickness, such as one half inch or less whereby the ultraviolet light emanating from lamp 19 is highly concentrated in the water flowing through chamber 18 rather than such ultraviolet light being diffused and require to pass through a greater depth of water before reaching the surrounding cylinder 11 as would occur if the cylinder 11 were substantially larger. For example, in many conventional ultraviolet water purification units of this type the annular water chamber is approximately one inch thick radially. The small radial dimension of the annular oxidation chamber 18 maintains the intensity of the 185 nm light at a substantial level throughout the radial depth of the water within the water chamber thus allowing substantial oxidation of organic material despite absorption of the 185 nm light by the water.

The ultraviolet light lamp 19 is preferably of a type to produce a significant portion of substantially 185nm wavelengths which is particularly suitable for producing the photodecomposition of the organics in the water although it is to be understood that the present invention is not limited to and does not exclude the use of ultraviolet light of different wavelengths that will produce adequate photodecomposition of the organics in water. The use of ultraviolet light in this range for analyzing the TOC in water in complex testing equipment is well known from U.S. patent No. 3,958,941. As noted previously, the photochemical oxidization of organic material in water by the use of ultraviolet light produces primarily carbon dioxide in the water which is harmless in many experiments and industrial processes.

The apparatus shown in Figs. 1 and 2 uses a standard length ultraviolet lamp of approximately 30" but it is to be understood that different lengths and configurations of lamps could be used, or multiple lamps could be used in a single vessel of a larger diameter. Also for more rapid or intense water treatment, two or more units can be connected in series in a system. Further, the 30" lamp 19 that has been used in experiments is a low pressure type using 40 watts of power that it is believed to produce about 13.8 ultraviolet watts and only about 5% of 185nm light and 95% of 254nm light, whereas it would be preferable that a greater percentage of 185nm light be produced and it is believed that such a lamp can be developed. Also, applicant is aware of high pressure and medium

pressure ultraviolet light lamps as produced by Hanovia Ltd. (HK), Phillips B.V. (Holland), Brown Bovari S.A. (Switzerland and others, that are currently used for bacteria control in Europe and The United Kingdom, which lamps use substantially higher wattage, for example 1850 watts of power, and produce substantially higher ultraviolet light output that may be useful in the present invention. With higher levels of output of 185nm ultraviolet light it is anticipated, although it has not been tested by applicant as yet, that the acceptable depth of water in the treatment chamber can be substantially larger than the 1/2" radial depth described above without sacrificing the intensity desired for effective operation of this invention.

→ The presence of oxygen dissolved in the water is believed to influence the production of the OH free radical and various other highly reactive chemical radical species which affect substantial oxidation of organic carbon to primarily carbon dioxide in the process performed by this invention. Thus, it is believed that any step in the process that can beneficially influence the production of such reactive chemical radical species will advantageously influence the process of the present invention.

It has also been found that the presence of certain material in contact with the water during the oxidation process can also substantially enhance the organic carbon oxidation process. One such material is titanium whereby making the cylinder 11 of titanium or lining it with titanium has been found to enhance the oxidation of the organic carbons to primarily carbon dioxide.

Referring now to Fig. 3, a system is illustrated for conditioning or reconditioning ion-exchange resin columns by the use of the TOC reduction unit 10 illustrated in Figs. 1 and 2. Two ion exchange resin columns 30 and 31 are illustrated as being connected in parallel between an inlet conduit 32 and an outlet conduit 33 but the system is equally applicable for use with a single resin column, or more than two resin columns, or connected in series between the conduits 32 and 33. A pump 34 is provided for circulating water from outlet conduit 33 to inlet pipe 12 on the unit 10 and inlet conduit 32 is connected from outlet pipe 13 of the unit 10 to the resin columns 30 and 31. A water supply pipe 35 is preferably connected through a pressure regulator 36 to pipe 32 for supplying deionized water to the system for the start-up of the conditioning or reconditioning cycle for the resin columns 30 and 31 after all of the piping connections have been made. With the ultraviolet light lamp 19 illuminated, the water is circulated continuously and repeatedly through the resin columns 30, 31 and the TOC reduction unit 10 to rinse organic material from the resin columns 30, 31 for passing through the unit

10 to photochemically oxidize the organics to primarily carbon dioxide which dissolves in the water and is returned to and removed by the resin in the resin columns 30 and 31. Other products in addition to carbon dioxide also may be formed by the photochemical oxidation. These other products are also removed by the ion-exchange resin in the resin columns 30 and 31. This recirculation of water is continued until a sufficiently low level of TOC in the resin columns is achieved. Organic material is found even in new resin columns in moderate amounts and can be substantially reduced or eliminated by this system. Organic impurities (TOC) may be found in very substantial quantities in a used resin column to the degree that the TOC adversely affects or fouls the resin and prevents proper deionization but the TOC may be substantially reduced and virtually eliminated by this ultraviolet reconditioning system.

20 * For example, tests have been conducted with the TOC reduction unit 10 of the above described construction shown in Figs. 1 and 2 used in a system as illustrated in Fig. 3 having five newly regenerated deionization resin columns connected in parallel. In one such test employing a water flow rate of one and one-half gallons per minute per cubic foot of resin the TOC was reduced from a starting level of approximately 700ppb (parts per billion) to approximately 400ppb (parts per billion) in the first hour of water recirculation, to approximately 300ppb in the second hour, to approximately 15 ppb in the next fourteen hours, and finally to the virtually negligible level of 1.5ppb after thirty-eight hours. In another similar test of reconditioning five deionization tanks connected in parallel in this system at a slower rate of one gallon per minute per cubic foot of resin, similar results were achieved in reducing the organics from approximately 750ppb to 400ppb in the first hour, to 250ppb in the next two hours, to a level of about 14ppb after a total of sixteen hours and finally to a level of 2.7ppb after a total of forty hours.

Referring again to Fig. 3, appropriate instrumentation may be provided for monitoring the operation of this system to determine when the TOC is of a sufficiently low level for the desired purposes. A resistivity meter 37 may be provided on the inlet pipe 12 of the unit 10 by mounting the meter on the aforedescribed branch conduit 15. A second resistivity meter 38 may be similarly mounted on the outlet pipe 13. In this manner the resistivity of the water may be measured at the inlet and outlet of the unit 10 to monitor the changes in resistivity of the water, which in turn provides an indication of dissolved carbon dioxide that can be related to the TOC, although not as a direct reading. Further, a TOC monitor 39 may be installed in conduit 32 for monitoring the actual

TOC which is preferable but such meters are substantially more expensive than resistivity meters. Thus, it is believed that appropriate monitoring by the less expensive resistivity meters 37 and 38 should be adequate for most systems.

In addition to the use of the TOC reduction unit 10 illustrated in Fig. 3 for regenerating or conditioning resin columns, it is contemplated that the unit 10 will also be advantageously usable in water supply systems for experiments and industrial processes wherein the unit 10 is installed in series in the water supply line with the inlet pipe 12 connected to the water supply and the outlet 13 connected to the water distribution line for substantially reducing the TOC in the supply water. For example, in present systems for supplying high purity water, there are provided filtration units for removing particulate matter and ion exchange columns for deionizing the water wherein the TOC reduction unit 10 may be installed in series, either upstream or downstream, with the filtration or deionization units for further improving the water quality by minimizing the TOC. The resultant water may have a quantity of carbon dioxide resulting from the photochemical decomposition of the organics but the carbon dioxide will not be objectionable in many water uses.

Referring now to Fig. 4, a typical water supply system for an industrial process is illustrated with the apparatus of the present invention employed at two different locations for advantageous improving the water supply system. The water is first introduced from any conventional source 40 into a prefiltration unit 41 for removing particulate matter and then flows through a conventional ultraviolet sterilization unit 42 (that uses 254nm ultraviolet light) into a reverse osmosis purification unit 43. Normally, the water is then passed through a deionization unit to a storage tank 44 where the water is stored for use. In the system of this invention two identical deionization units 45 and 46 are provided and connected in parallel with each other with appropriate valving for flow water through one or the other of the units 45 or 46 at one time. A TOC reduction unit 10a of this invention is connected to the deionization units 45 and 46 and a pump 47 is provided for selectively circulating water through the TOC reduction unit 10a and one of the deionization units 45 or 46. For example, by opening valves 48 and 52 and closing valves 50 and 54, the water from reverse osmosis unit 43 flows only through deionization unit 45 to the storage tank 44. Further, by closing valves 49 and 53 and opening valves 51 and 55, water can be circulated by pump 47 through TOC reduction unit 10a and deionization unit 46 to reduce the organics in unit 46, similar to the system of Fig. 3. Thus, by periodically switching the positions of the

valves 48-55 to and from the aforesaid positions, one deionization unit can be in use for deionizing the water while the other deionization unit is being treated to reduce the organics.

Further with respect to Fig. 4, the clean water from storage tank 44 is then supplied through a multiple column deionization unit 56, another 254nm ultraviolet sterilizing unit 57, and a sub-micron filter 58 to the point of use 59 in the industrial process which, for example, may be the washing of computer chip components of the like. The used water is then conducted to a reclaim tank 60 where its purity can be tested and monitored to determine whether it is sufficiently clean to allow reprocessing and reuse or if it must be discarded. If the used water is sufficiently clean to permit reuse, it is supplied through a carbon filtration unit 61, a deionization unit 62, and another sterilizing unit 63 to the storage tank 44. In order to maximize the quality of the water in the reclaim tank 60 to permit its reuse a recirculating system is provided and includes a deionization unit 64, a TOC reduction unit 10b, and a pump 65 whereby the water is continually deionized in unit 64 and the organics in unit 64 and the water are maintained at a low level by the TOC reduction unit 10b of this invention. By this system the maximum use and reuse of the water is possible.

Although the present invention has been described in connection with a preferred embodiment of the TOC reduction unit and two types of preferred systems within which the unit is used, it will readily appear to those skilled in the art and is to be understood that the present invention may take many forms and be applicable to various systems within the scope of the appended claims.

Claims

1. An organic reduction apparatus for conditioning water containing organic material, comprising, an oxidation chamber having an ultraviolet lamp for producing 185nm ultraviolet light, and said oxidation chamber having means for conducting a layer of water past said ultraviolet lamp for causing intense exposure to the ultraviolet light for photochemically oxidizing the organic material to form primarily carbon dioxide.

2. The apparatus of claim 1 wherein said oxidation chamber includes an elongated cylinder with a quartz sleeve positioned concentrically within the cylinder to form a thin annular passage for the water between the cylinder and sleeve, and said ultraviolet lamp positioned inside said sleeve.

3. An organic reduction apparatus for conditioning ion exchange resin in a vessel having an inlet and an outlet, comprising, an oxidation chamber

having an ultraviolet lamp for producing 185 nm ultraviolet light, and means for connecting said oxidation chamber between the vessel inlet and outlet and for causing circulation of water from the vessel outlet to and through the oxidation chamber for exposure to the ultraviolet light and back to the vessel inlet.

4. The apparatus of claim 3 wherein means are provided for measuring and monitoring a characteristic of the circulating water for determining the level of organic material in the water.

5. The apparatus of claim 4 wherein said oxidation chamber is comprised of a cylinder with a transparent sleeve therein for forming an annular passage therebetween for the water, and the ultraviolet lamp is positioned within said sleeve.

6. The apparatus of claim 2 or 5 wherein said annular passage is of a radial thickness from the sleeve to the cylinder of approximately one-half inch or less for causing said intense exposure to ultraviolet light.

7. The apparatus of claim 6 wherein said ultraviolet lamp is approximately 30" long and has an output of about 13.8 ultraviolet watts.

8. The apparatus of any one or claims 1 to 4 wherein the ultraviolet light intensity is at least equal to the level produced by a 30" long ultraviolet lamp in the center of a cylindrical chamber with a $\frac{1}{2}$ " layer of water surrounding the lamp, which lamp produces 13.8 ultraviolet watts.

9. The apparatus of any preceding claim wherein said oxidation chamber includes a substantial surface area comprised of titanium and in contact with the water.

10. An apparatus for conditioning organic carbon contaminated ion exchange resin in a vessel having an inlet and an outlet, comprising, a cylindrical body, a quartz sleeve extending the length of and concentrically positioned inside said cylindrical body to form a narrow annular passage between the outside of the quartz sleeve and the inside of the cylindrical body, said cylindrical body having an inlet on the outer surface at one end and an outlet at the other end communicating with said annular passage for conducting water from said one end to the other end through said passage, an ultraviolet lamp for producing 185nm ultraviolet light mounted within said quartz sleeve and extending the length of said cylindrical body for exposing the water in said annular passage to the ultraviolet light, means for connecting the vessel outlet to the cylindrical body inlet and the cylindrical body outlet to the vessel inlet including pump means for repeatedly circulating the water from the vessel through the annular passage and back to the vessel for repeated exposure of the water and organic carbon contamination rinsed from the resin to the ultraviolet light for oxidizing the organic carbon

contamination to primarily carbon dioxide which is in turn removed from the circulating water by the resin.

11. The apparatus of claim 10 wherein said narrow annular passage is approximately one-half inch thick or less in the radial direction between the outside of quartz sleeve and the inside of the cylindrical body, and said ultraviolet lamp is approximately 30" long and produces ultraviolet watts.

12. A method of conditioning organic carbon contaminated water comprising the steps of recirculating the contaminated water past and exposing the water to intense levels of 185nm ultraviolet light for oxidizing the organic carbon to primarily carbon dioxide.

13. A method of conditioning organic carbon contaminated ion exchange resin in a vessel, comprising the steps of, circulating water through the vessel past the resin to entrain organic carbon in the water, circulating and exposing the water and entrained organic carbon to intense levels of 185nm ultraviolet light for oxidizing the organic carbon to primarily carbon dioxide, circulating the water and carbon dioxide back to the vessel for entraining more organic carbon in the water, and continuing said circulation of water to and from the vessel until the level of organic carbon entrained in the water passing from the vessel is at a predetermined desired level.

14. The method of claim 13 including the step of removing the carbon dioxide in the vessel by adsorption by the resin.

15. The method of claim 13 or 14 including monitoring the TOC level of the water for determining when said predetermined desired level has been achieved.

16. The method of claim 13 or 14 including monitoring the resistivity of the water for determining when said predetermined desired level has been achieved.

17. The method of any one of claims 12 to 16 including the step of exposing the water to a substantial surface area of titanium during the step of exposing the water to ultraviolet light.

18. A system for conditioning organic carbon contaminated ion exchange resin in vessel in a water system, comprising, selectively isolating a first ion exchange resin vessel from the water supply system while retaining a second ion exchange resin vessel in operative relationship in the water system, connecting the first ion exchange resin vessel in a recirculation system for recirculating water therethrough and exposing the water to 185nm ultraviolet light for oxidation of the organic carbon, and alternately selectively isolating the second ion exchange resin vessel from the water system while retaining the first ion exchange resin

vessel in the water supply system and connecting the second ion exchange resin vessel to the recirculation system.

19. A system for conditioning organic carbon contaminated ion exchange resin in vessel in a water system, comprising, first and second vessels containing ion exchange resin, means connecting said first and second vessels in parallel in the water system, a recirculating system connected to said first and second vessels and including a pump for circulating water and means for exposing the circulating water to 185nm ultraviolet light for oxidation of the organic carbon, and valve means for selectively isolating one of said first and second vessels from the water system and connecting that isolated vessel to the recirculating system while the other of said first and second vessels remains connected in the water system.

20. A system for conditioning reclaimed water in tank of a water system, comprising, a water recirculating system for taking water from and returning water to the tank, a deionization means in said recirculating system, and said recirculating system including means for exposing the water circulated therethrough to 185nm ultraviolet light to oxidize any organic carbon in the water.

21. The system of claim 20 wherein the recirculating system passes the water from the tank through the deionization means first and then through said means for exposing the water to 185nm ultraviolet light.

22. A system for protecting ion exchange resin in a vessel from contamination due to organic carbon in supply water, comprising, means connected upstream of the vessel for receiving all of the supply water before such water reaches the vessel, and said means including means for exposing the water to intense 185nm ultraviolet light.

23. The system of any one of claims 18 to 22 wherein the recirculating system includes a substantial surface area of titanium in contact with the water.

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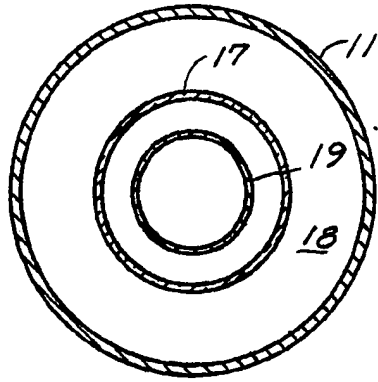


FIG. 2.

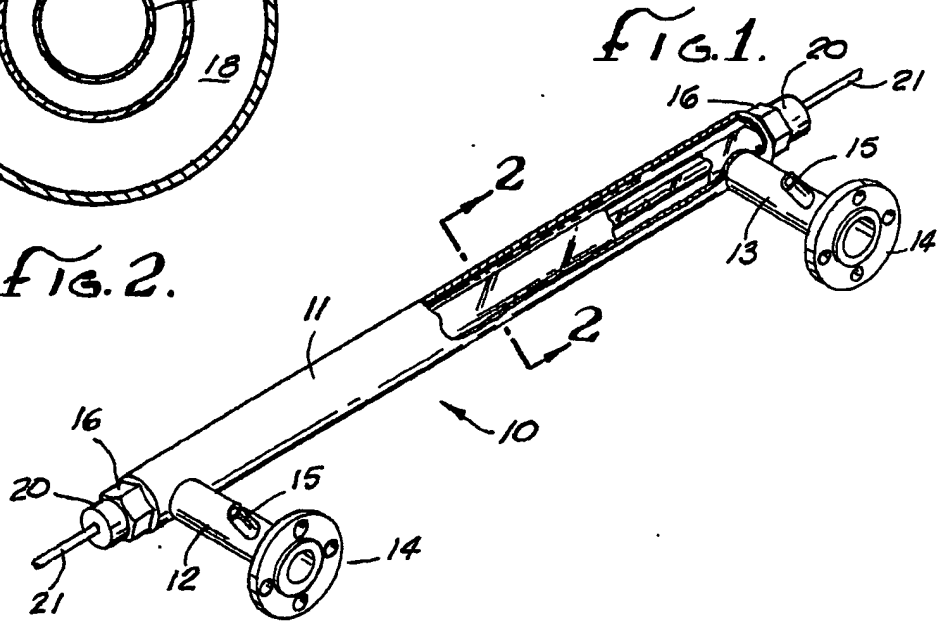


FIG. 1.

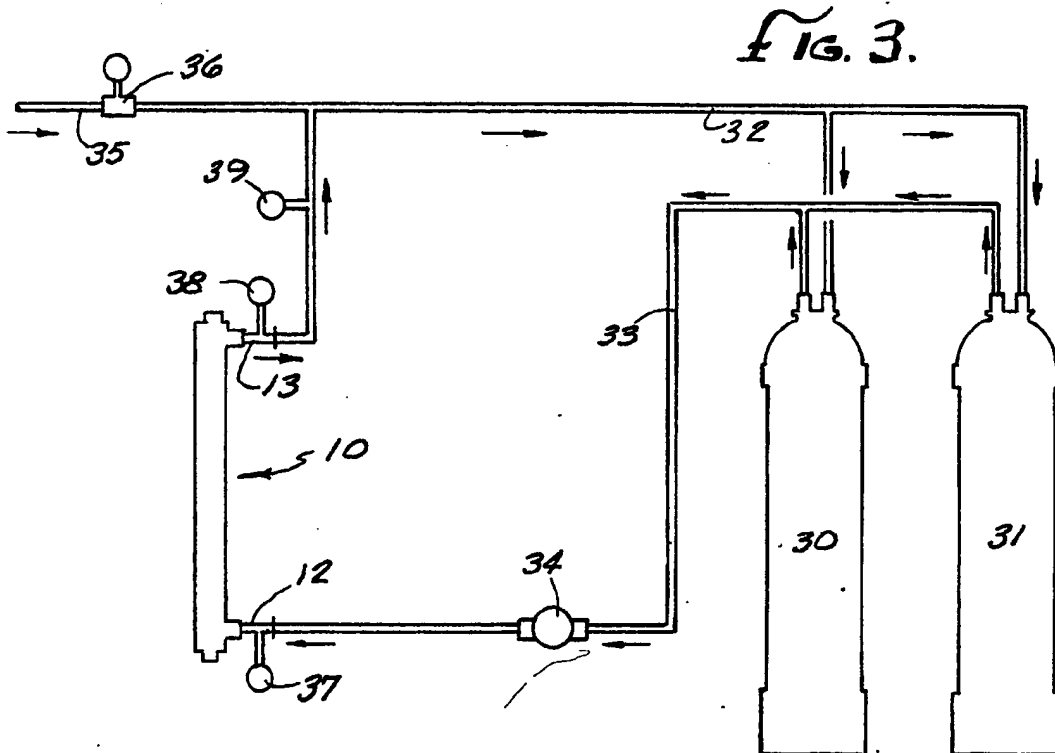


FIG. 3.

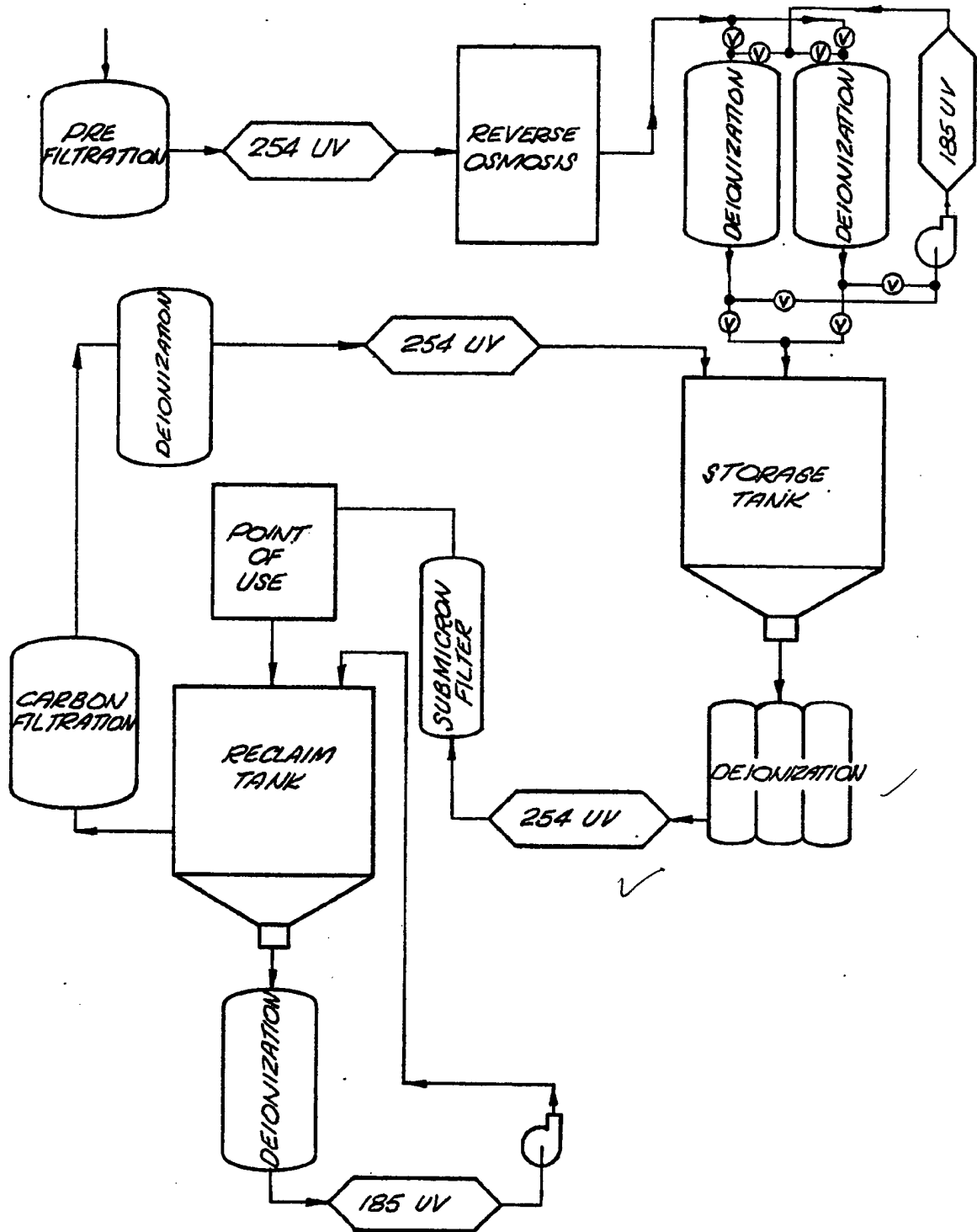


FIG. 4.



European Patent
Office

EUROPEAN SEARCH REPORT

Application number

DOCUMENTS CONSIDERED TO BE RELEVANT			EP 87302476.4
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
D, Y	<u>US - A - 3 958 941 (REGAN)</u> * Column 1, lines 25-28 * --	1	C 02 F 1/32 B 01 J 49/00
Y	<u>DE - A1 - 3 020 170 (H. & P. KAAS)</u> * Claims 2,4 * --	1	
A	<u>DE - A - 1 937 126 (RAUH)</u> * Page 6, line 1 - page 7, line 8 * ----	1,2	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			C 02 F B 01 J G 01 N 31/00
The present search report has been drawn up for all claims			
Place of search VIENNA		Date of completion of the search 23-12-1987	Examiner WILFLINGER
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	